

# Appendix A: Theory of Fission Chains and Count Distributions

**Contains Appendix A of UCRL-TR-217147** 

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U.S. Department of Energy

Lawrence Livermore National Laboratory **September 13, 2005** 

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This work was performed under the auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

## **Appendix A**

### **Theory of Fission Chains and Count Distributions**

#### **Poisson counting distribution**

A random source produces a Poisson counting distribution,

$$P_n(T) = \frac{\overline{C}^n}{n!} e^{-\overline{C}}$$
 A1

for the probability to count n neutrons in time T, where  $\overline{C} = R$  T is the average number of counts during that counting time T, and R is the count rate. This formula is the foundation from which deviations indicate the presence of fission. Because of its importance we present a derivation. The method of derivation is to construct a time evolution equation for the number of counts obtained up to a given time. Let  $P_n(t + \Delta t)$  be the probability to count n neutrons by time  $t + \Delta t$ . This probability will be related to processes that can occur between times t and  $t + \Delta t$ . The equation is

$$P_n(t + \Delta t) = P_n(t)(1 - R\Delta t) + P_{n-1}(t)R\Delta t$$
 A2

The probability of getting a count in any infinitesimal time interval  $\Delta t$  is R  $\Delta t$ . This is independent of any independent of any previous history, so any  $\Delta t$  has the same probability to get a count given that the count rate is R. The meaning of the first term on the right hand side is that there were n counts as of time t, and no counts were obtained between times t and  $t + \Delta t$ . This occurs with the factor  $(1 - R \Delta t)$ , the probability that there was no count. Consequently if the number of counts recorded between time zero and time t was n, this number does not change in going from time t to time  $t + \Delta t$ . The second term on the right hand side is the probability that as of time t there were only n-1 counts, and between time t and  $t + \Delta t$  a count was obtained. Because  $\Delta t$  is infinitesimal, there is no chance to get more than one count in  $\Delta t$ . This rate equation then becomes a differential equation as  $\Delta t \rightarrow 0$ ,

$$\frac{\partial P_n(t)}{\partial t} = -R P_n(t) + P_{n-1}(t)R$$
 A3

It is convenient to construct a generating function for the probability distribution by multiplying  $P_n(t)$  by  $x^n$ , and summing over n,  $\pi(x,t) = \sum_{n=0}^{\infty} P_n(t) \chi^n$ . Multiplying Equation A3 by  $x^n$  gives

$$\frac{\partial}{\partial t} \sum_{n=0}^{\infty} P_n(t) \chi^n = -R \sum_{n=0}^{\infty} P_n(t) \chi^n + R \sum_{n=1}^{\infty} P_{n-1}(t) \chi^{n-1} \chi$$
 A4

In the last term the sum begins at n = 1 because the smallest value on n is 0. This gives an equation for  $\pi(x,t)$ ,

$$\frac{\partial}{\partial t}\pi = -R\pi + Rx\pi = R(x-1)\pi$$
 A5

The solution of this differential equation, with the initial condition  $\pi(x, t = 0) = 1$ , since at the beginning of counting at t = 0 no neutrons were counted, is

$$\pi(x,t) = e^{Rt(x-1)} = \sum_{n=0}^{\infty} \frac{(Rt)^n}{n!} e^{-Rt} x^n$$
 A6

Using this generating function the moments of the counting distribution are obtained by differentiation with respect to x, and then setting x = 1. The first moment is

$$\frac{\partial}{\partial x}\pi = \sum_{n=1}^{\infty} n \frac{\left(Rt\right)^n}{n!} e^{-Rt} \chi^{n-1} = Rt e^{Rt(x-1)}$$
 A7

Setting x = 1, gives

$$\overline{C} = \sum_{n=1}^{\infty} n \frac{(Rt)^n}{n!} e^{-Rt} = Rt$$

The second derivative divided by 2 gives the average number of pairs,

$$\frac{1}{2} \frac{\partial^2}{\partial x^2} \pi = \sum_{n=1}^{\infty} \frac{n(n-1)}{2} \frac{(Rt)^n}{n!} e^{-Rt} x^{n-2} = \frac{(Rt)^2}{2} e^{Rt(x-1)}$$
 A9

Setting x = 1 gives the pair moment,

$$\sum_{n=3}^{\infty} \frac{n(n-1)}{2} \frac{(Rt)^n}{n!} e^{-Rt} = \frac{(Rt)^n}{2}$$
 A10

Also, from the pair moment and the mean, the variance can be obtained, since

$$2\sum_{n=2}^{\infty} \frac{n(n-1)}{2} \frac{(Rt)^n}{n!} e^{-Rt} + \sum_{n=1}^{\infty} n \frac{(Rt)^n}{n!} e^{-Rt} = \sum_{n=2}^{\infty} n^2 \frac{(Rt)^n}{n!} e^{-Rt} = 2\frac{(Rt)^n}{2} + Rt$$
A11

and so, from Equations A8 and A12, the variance is

$$\left\langle \left( n - \overline{C} \right) \right\rangle = \sum_{n=2}^{\infty} n^2 \frac{\left( Rt \right)^n}{n!} e^{-Rt} - \left[ \sum_{n=1}^{\infty} n \frac{\left( Rt \right)^n}{n!} e^{-Rt} \right]^2 = Rt$$
 A12

that is, the variance of a Poisson distribution is equal to its mean. This completes the derivation of some of the results stated in the text in the discussion of Feynman variance [1].

Of particular importance is the formula for no counts in a random time interval,  $P_0(t) = e^{-Rt}$ . The probability to get a count in an infinitesimal random time interval is  $R \Delta t$ , the probability to observe for time t getting no counts, and then getting a count between t and  $t + \Delta t$  is  $e^{-Rt} R \Delta t$ . This probability is largest for small t. The probability to get a count at some time is

$$\int_{0}^{\infty} e^{-Rt} R dt = 1$$
 A13

and the average time to get a count after randomly observing is

$$\int_{0}^{\infty} t \, e^{-Rt} \, R dt = R^{-1} \tag{A14}$$

These properties of a random process will be used below.

#### Counting distribution from fission source

The form of the counting distribution from a fission source is a generalized Poisson distribution [5]. While the Poisson distribution is characterized by a single parameter, the count rate R, the generalized Poisson distribution depends on many, in principle even an infinite number, of time dependent functions,  $\Lambda_k(T)$ , k = 1, 2, 3, ..., which have the meaning of the probability to count k neutrons from the *same* fission chain in the time gate T. These functions are characterized by a finite number of parameters. If  $b_n(T)$  is the probability to count n neutrons in a random time gate of duration T, then,

$$b_{n} = b_{0} \sum_{i_{1}+2i_{2}+3i_{3}+\cdots ni_{n}=n} \frac{\Lambda_{1}^{i_{1}} \Lambda_{2}^{i_{2}} \cdots \Lambda_{n}^{i_{n}}}{i_{1}! i_{2}! \cdots i_{n}!}$$
 A15

where  $i_k$  is the number of independent chains contributing k counts (for k = n,  $i_n = 0$  or 1, while for k = 1,  $i_1 = 0,1,2,\dots,n$ ), and

$$b_0 = e^{-\left(\Lambda_1 + \Lambda_2 + \dots + \Lambda_n + \dots\right)}$$
 A16

For example, the probability to get 5 counts is

$$b_{5} = \left(\Lambda_{5} + \Lambda_{4}\Lambda_{1} + \Lambda_{3}\Lambda_{2} + \Lambda_{3}\frac{\Lambda_{1}^{2}}{2} + \frac{\Lambda_{2}^{2}}{2}\Lambda_{1} + \Lambda_{2}\frac{\Lambda_{1}^{3}}{3!} + \frac{\Lambda_{1}^{5}}{5!}\right)b_{0}$$
 A17

If all the  $\Lambda_k(T)$  but  $\Lambda_1$  are zero, then  $b_5 \rightarrow \frac{\Lambda_1}{5!} e^{-\Lambda_1}$ , a Poisson distribution. The term  $\frac{\Lambda_1}{5!} b_0$  represents the probability that each of the 5 counts was due to an independent random source, where only a single neutron is counted from each independent chain. The term  $\Lambda_5 b_0$  is the probability that all 5 counts arise from a common source, a single chain. The term

 $\frac{\Lambda_2^2}{2}\Lambda_1 b_0$ , for example, is the probability that the 5 counted neutrons arise from 3 independent

random sources, two pairs of counts each have a different common ancestor, and an additional count arises from a third source. For a weak neutron source in a system of high multiplication, the chains are few and far between, but if there are n counts it is likely they are all from the same chain. For a strong source in a system of low multiplication, the probability of getting multiple counts from a single chain is small, while the probability of getting many counts, mostly from independent chains, is high. Information about the source strength and multiplication are encoded in the counting distribution.

The generating function for the generalized Poisson count distribution is [5],

$$\pi(z) = \sum_{n=0}^{\infty} b_n(T) z^n = \exp\left[\sum_{j=1}^{\infty} \left(z^j - 1\right) \Lambda_j(T)\right]$$
 A18

Expanding the generating function in powers of z gives Eq. (A16) for  $b(T)_n$  in terms of  $\Lambda_k$ . The probability of detecting j counts from a single chain within the time gate T is [5],

$$\Lambda_{j}(T) = F_{s} \sum_{v=j}^{\infty} P_{v} \sum_{n=1}^{\infty} \left[ \int_{n}^{v} e^{n} \left( 1 - \varepsilon \right)^{-n} \left\{ \int_{-\infty}^{0} ds \binom{n}{j} \left[ \int_{0}^{T} e^{-\lambda(t-s)} \lambda dt \right]^{j} \left[ 1 - \int_{0}^{T} e^{-\lambda(t-s)} \lambda dt \right]^{n-j} + \int_{0}^{T} ds \binom{n}{j} \left[ \int_{s}^{T} e^{-\lambda(t-s)} \lambda dt \right]^{j} \left[ 1 - \int_{s}^{T} e^{-\lambda(t-s)} \lambda dt \right]^{n-j} \right\}$$
A19

Physically  $\Lambda_i(T)$  is the probability to count j neutrons within a time gate between t = 0 and t = 0T. It is a product of probabilities: First there is the probability of spontaneous creation of a source neutron within a small time interval,  $F_s dt$ , where  $F_s$  is the rate of spontaneous fission. Then given a spontaneous neutron created between times s and s  $+\Delta s$ , a fission chain is assumed to be created instantaneously with probability  $P_{\nu}$  that  $\nu$  neutrons were created by the fission chain (after subtracting those neutrons absorbed by fissions). Out of the  $\nu$  neutrons created, n are detected, each with probability  $\varepsilon$  (efficiency). This is a binomial distribution, all the ways of detecting n neutrons out of  $\nu$ , each with probability  $\varepsilon$ , and not detecting with probability (1 -  $\varepsilon$ ) each the remaining (v - n) neutrons. Of the n detected neutrons, j are detected within the time gate between times t = 0 and t = T. A neutron created at time s has a probability,  $e^{-\lambda(t-s)}$  to survive until time t, and  $\lambda dt$  is the probability of being captured within the time dt. The probability of detecting j, out of the n detected neutrons, within the time gate of duration T is again a binomial distribution in the probability,  $e^{-\lambda(t-s)}\lambda dt$ . There are two time ordering terms, representing fission chains that take place before the counting window, and fission chains that are initiated within the counting window. This formula can be simplified, first by use of the identity for a product of binomial distributions,

$$\sum_{n=j}^{\nu} {v \choose n} \varepsilon^n (1-\varepsilon)^{-n} {n \choose j} x^j (1-x)^{\nu-j} = {v \choose j} (\varepsilon x)^j (1-\varepsilon x)^{\nu-j}$$
 A20

With this binomial simplification, then becomes,

$$\Lambda_{j} = F_{s} \int_{-\infty}^{0} \left[ \sum_{v=j}^{\infty} P_{v} {v \choose j} (\varepsilon x) (1 - \varepsilon x)^{-j} \right] ds$$

$$+ F_{s} \int_{0}^{T} \left[ \sum_{v=j}^{\infty} P_{v} {v \choose j} (\varepsilon y) (1 - \varepsilon y)^{-j} \right] ds$$
A21

where

$$x = \int_{0}^{T} e^{-\lambda(t-s)} \lambda dt = e^{\lambda s} \left(1 - e^{-\lambda T}\right)$$
 A22

and

$$y = \int_{s}^{T} e^{-\lambda(t-s)} \lambda dt = \left(1 - e^{-\lambda(T-s)}\right)$$
 A23

The  $\varepsilon x$  integral is the probability that a neutron created at  $\mathbf{s} < 0$  survives in the detector from time  $\mathbf{s} = 0$  until time t, and then is lost to the system between t and  $\mathbf{t} + d\mathbf{t}$ , but now  $\varepsilon$  is the fraction of those removed neutrons that are detected. The neutrons diffusing in the detector could have been lost to absorptions that did not result in counts, or lost to leakage. A similar interpretation is given for the  $\varepsilon y$  integral except that the fission chain takes place at time s during the counting window.

For the generating function, consider the sum:

$$\sum_{j=0}^{\infty} \Lambda_{j} z^{j} = F_{s} \int_{-\infty}^{0} \left[ \sum_{v=0}^{\infty} P_{v} \sum_{j=0}^{v} {v \choose j} (\varepsilon x z)^{j} (1 - \varepsilon x)^{-j} \right] ds$$

$$+ F_{s} \int_{0}^{T} \left[ \sum_{v=0}^{\infty} P_{v} \sum_{j=0}^{v} {v \choose j} (\varepsilon y z)^{j} (1 - \varepsilon y)^{-j} \right] ds$$

$$= F_{s} \int_{-\infty}^{0} \sum_{v=0}^{\infty} P_{v} \left[ \varepsilon x z + (1 - \varepsilon x) \right] ds + F_{s} \int_{0}^{T} \sum_{v=0}^{\infty} P_{v} \left[ \varepsilon y z + (1 - \varepsilon y) \right] ds$$

$$= F_{s} \int_{-\infty}^{0} \sum_{v=0}^{\infty} P_{v} \left[ 1 - \varepsilon (1 - z) (1 - e^{-\lambda T}) \right] ds + F_{s} \int_{0}^{T} \sum_{v=0}^{\infty} P_{v} \left[ 1 - \varepsilon (1 - z) (1 - e^{-\lambda (T - s)}) \right] ds \quad A21$$

In the second integral of the last line of Equation A24, the change of the integration variable: T - s = u, ds = -du, and for s = 0, u = T and for s = T, u = 0, gives

$$\int_{0}^{T} \left[ 1 - \varepsilon \left( 1 - z \right) \left( 1 - e^{-\lambda (T - s)} \right) \right] ds$$

$$= \int_{0}^{T} \left[ 1 - \varepsilon \left( 1 - z \right) \left( 1 - e^{-\lambda u} \right) \right] du$$
 A25

In the first integral of the last line of Equation A24, the substitution  $(1 - e^{-\lambda T})e^{\lambda s} = 1 - e^{-\lambda u}$ ,

$$ds = \frac{e^{-\pi u}}{\left(1 - e^{-\pi u}\right)} du$$
, and for  $s = 0$ ,  $u = T$  and for  $s = -\infty$ ,  $u = 0$ , gives

$$\int_{-\infty}^{0} \left[ 1 - \varepsilon \left( 1 - z \right) \left( 1 - e^{-\lambda T} \right) e^{\lambda s} \right] ds$$

$$\int_{0}^{T} \left[ 1 - \varepsilon \left( 1 - z \right) \left( 1 - e^{-\lambda u} \right) \right] \frac{e^{-\lambda u}}{\left( 1 - e^{-\lambda u} \right)} du$$
A26

The sum gives

$$\sum_{j=0}^{\infty} \Lambda_{j} z^{j} = F_{s} \int_{0}^{T} \frac{1}{\left(1 - e^{-\lambda t}\right)} \sum_{r=0}^{\infty} P_{r} \left[1 - \varepsilon \left(1 - z\right) \left(1 - e^{-\lambda t}\right)\right] dt$$
 A27

where we substituted back the dummy integration variable t for u. This expression has the form of an integral over a generating function for the fission chain,

$$h[z] = \sum_{v=0}^{\infty} P_v z^v$$
 A28

that is,

$$\sum_{j=0}^{\infty} \Lambda_{j} z^{j} = F_{s} \int_{0}^{T} \frac{1}{(1-e^{-\lambda t})^{n}} \left[ 1 - \varepsilon (1-z) (1-e^{-\lambda t}) \right] t$$
 A29

Since  $P_v$  is a normalized probability distribution, h[1] = 1, and so the j = 0 terms cancel in the combination,

$$\sum_{j=1}^{\infty} \Lambda_{j} \left( z^{j} - 1 \right) = F_{s} \int_{0}^{T} \frac{1}{\left( -e^{-\lambda t} \right)} \left\{ \left( \left[ -e^{-\lambda t} \right] \right) \left( \left[ -e^{-\lambda t} \right] \right) \right\} dt$$
 A30

The generating function for the counting distribution is [9] then, from Equation A18,

$$\pi(z,T) = \sum_{n=0}^{\infty} b_n(T) z^n = \exp \left[ F \int_0^T \frac{1}{(-e^{-\lambda t})} \left[ \left[ -\varepsilon (1-z) \left( -e^{-\lambda t} \right) \right] \right] dt \right]$$
 A31

Consequently, the time dependent counting distribution can be obtained from the fission chain generating function. In this expression the time dependence is associated only with neutron diffusion, and appears in the generating function variable of the fission chain counting distribution. In the next section we will find a general series expression for this fission chain generating function.

#### Theory of fission chain distributions

#### Rate equation for time dependent internal neutron population

In order to compute  $\Lambda_j$ , the probabilities from which the count distribution is constructed through Equations A30 and A31, we must determine the probability  $P_v$  that a fission chain produces v neutrons that are not absorbed in producing subsequent fissions. This distribution depends on the probability p that a fission neutron induces a subsequent fission, and on the probability distribution for the number of neutrons produced by an individual fission process. We begin with the rate equation for the number of neutrons in the system as a function of time, starting from a single neutron at t = 0. For a subcritical system, this number distribution will go to zero as  $t \to \infty$ ,  $P_0(t) \to 1$ . We are interested in the number of neutrons that are available for detection. As our detector is external to the multiplying medium, this is the number or neutrons that leaks out of the system. As the chain evolves, some of the produced neutrons may produce subsequent fissions, perpetuating the chain, and other neutrons may be absorbed or leak. A model for the time dependent neutron population, in the approximation of neglecting spatial dependence and neutron energy dependence, was studied by Feynman [1]. The probability that there are k neutrons in the system at time  $t + \Delta t$  is determined from the number at time t by the rate equation [1 and 4]:

$$P_{k}(t + \Delta t) = P_{k}(t)\left(1 - k\frac{\Delta t}{\tau}\right) + qP_{k+1}(t)(k+1)\frac{\Delta t}{\tau} + p\sum_{v=0} P_{k-v+1}(t)C_{v}(k-v+1)\frac{\Delta t}{\tau}$$
 A32

The first term on the right hand side of the equation is the probability that none of the k neutrons in the system had an interaction within  $\Delta t$  that would change the number of neutrons,  $\tau$  being the neutron lifetime against any absorption, such as  $(n, \gamma)$  or (n, f), or leakage. Each of the neutrons has a probability  $\left(1 - \frac{\Delta t}{\tau}\right)$  of not interacting, so the term given is a linearization of

$$\left(1-\frac{\Delta t}{\tau}\right)^k$$
. The second term is the probability that there were k + 1 neutrons at time t, and one

of them was absorbed without creating another neutron, or leaked. If p is the probability that a neutron interaction gives fission, q = 1 - p is the probability for non-fission absorption or leakage. The final term gives the probability that between t and  $t + \Delta t$  an interaction took place,

 $\frac{\Delta t}{\tau}$ , with probability p that the interaction produced fission. From the original k + 1 - v neutrons

at time t, one neutron was absorbed, and with probability  $C_{\nu}$ ,  $\nu$  neutrons were created in the induced fission. This  $C_{\nu}$  distribution is input nuclear data, with maximum number of emitted neutrons in a fission induced with fission spectrum neutrons typically about 8. This equation can be solved using a probability generating function [1 and 4],

$$f(t,x) = \sum_{k=0}^{\infty} P_k(t) \chi^k$$
 A33

In the limit  $\Delta t \rightarrow 0$ , a differential equation for the generating function is,

$$\tau \frac{\partial f}{\partial t} = g(x) \frac{\partial f}{\partial x}$$
 A34

where,

$$g(x) = -x + q + pC(x)$$
 A35

and,

$$C(x) = \sum_{v=0}^{\infty} C_v x^v$$
 A36

The equation says that differentiating f(t, x) with respect to t/t is equivalent to differentiating f with respect to

$$G(x) = \int \frac{dx}{g(x)}$$
 A37

that is, f(t, x) is a function of  $\left[\frac{t}{\tau} + G(x)\right]$ . Since for  $t \to 0$ , there is one neutron in the system, f(0, x) = x, or  $P_1(0) = 1$ , and all other  $P_k(0) = 0$ . This implies [1] the arbitrary function of  $\left[\frac{t}{\tau} + G(x)\right]$  is  $G^{-1}$ ,

$$G[f(t,x)] = \frac{t}{\tau} + G(x)$$
 A38

If  $G^{-1}$  can be found, then this equation constitutes the solution to the time dependent neutron population. As  $t \to \infty$ ,  $G[f(t,x)] \to \infty$ . This implies that if  $f(t,x) \to h(x)$  as,  $t \to \infty$ , then since

$$G[h(x)] = \int \frac{dx}{g[h(x)]} \to \infty$$
 A39

this is realized by,

$$g[h(x)] = 0 A40$$

This condition is, from Equation A35,

$$g[h(x)] = 0 = -h(x) + q + p \sum_{v=0} C_v [h(x)]$$
 A41

which has a solution in the *supercritical* case (see below for a discussion of supercritical) for h(x) = Q = constant, independent of x. So  $f(t,x) \to Q$  as  $t \to \infty$ , which has the interpretation of the probability that there are no neutrons in the supercritical system as  $t \to \infty$ .

#### Time dependence

The rate of approach to the asymptotic value in time depends on the time evolution of the chain. The partial differential equation for the time dependent generating function, Equation A34, has a time scale  $\tau$ , the average time for the neutron to undergo a non-scattering interaction or leakage. The time evolution of a fission chain depends not only on  $\tau$  but also on the amount of

multiplication, that is, the length of the chain. The multiplication is defined,  $M = \frac{1}{(1-k_{eff})}$ ,

where in this model  $K_{eff} = vp$ , where  $v = \sum_{v=1}^{\infty} v C_v$  is the average number of neutron created by a single induced fission. For example, the first moment of the time evolution equation is,

$$\tau \frac{\partial^2 f}{\partial t \partial x} = g'(x) \frac{\partial f}{\partial x} + g(x) \frac{\partial^2 f}{\partial x^2}$$
 A42

where

$$g'(x) = -1 + q + pC'(x)$$
 A43

and where

$$C'(x) = \sum_{v=0} v C_v x^{v-1}$$
 A44

Setting x = 1, Equation A44 gives v, and with Equation A33, A42 and 43, and also using g(1) = 0 from Equation A35 since C(1) = 1), the first moment equation,

$$\tau \frac{\partial}{\partial t} \sum_{k=1}^{\infty} k P_k(t) = \left( 1 - vp \right) \sum_{k=1}^{\infty} k P_k(t)$$
 A45

The solution with the initial condition that there was one neutron in the system at t = 0 is

$$\sum_{k=1}^{\infty} k P_k(t) = e^{-\frac{\left(1-v_p\right)^2}{\tau}}$$
 A46

It is found that generally the time dependence is a function of  $e^{-\alpha t}$ , where  $\alpha = \frac{(1-vp)}{\tau}$ . When  $\alpha$ 

> 0, the system is *subcritical*, and when  $\alpha$  < 0, the system is *supercritical*, the average number of neutrons increases exponentially in time. The system is *critical* for  $k_{eff} = 1$ . For subcritical systems of current interest  $\alpha$  > 0, and the number of neutrons in the system will always go to zero for  $t >> \alpha^{-1}$ . The value for  $\tau$  in multiplying HEU or Pu metal is less than 1 shake (where a shake =  $10^{-8}$  seconds). This follows from the nuclear data for interactions. For fission spectrum neutron energies of order 1 MeV, the interaction cross sections are a few barns, the nuclei number density is of order 1/20 per barn-cm ( $\rho$  N<sub>0</sub>/A  $\sim$  20 gm/cm<sup>3</sup> x 0.6 x  $10^{24}$ / 240 gm), so the interaction mean free path is less than 20 cm. (The neutron leakage distance for a subcritical metal system is perhaps of order the scattering mean free path, a few cm for scattering cross sections of several barns.) The speed of a 1 MeV neutron is about 14 cm/shake, so the typical time scale for  $\tau$  is of order 1 shake. For multiplication of order 10, this gives  $\alpha^{-1}$  of order 10 shakes. The long time asymptotic fission chain distributions are therefore typically approached in much less than the 1  $\mu$ s.

In our discussion above of the count distribution we considered only a single time scale  $\lambda^{-1}$  characteristic of neutron diffusion through hydrogenous material, of order 10's or 100's of  $\mu$ s. One of the assumptions we make in the theoretical model is therefore that  $\chi^{-1} >> \chi^{-1}$ . (For thermal reactor problems, usually the inequality is reversed, the fission chains evolve on a very

long time scale. In that case many of the formulas below need to be corrected. Quite remarkably the equations for the theory discussed below simplify in the thermal reactor regime, and we have closed form analytic formulas for the time evolving fission chain populations and for the count distribution in this extreme regime [16].

The meaning of  $e^{-\alpha t}$  is also the probability that a neutron created at t=0 survives until time t. In the absence of fission, this probability is just,  $e^{-\frac{t}{\tau}}$ , the decay probability being determined by the total neutron lifetime. With fission, the neutron population is replenished, the survival probability decaying slower. The probable number of neutrons at time t, starting from one neutron at t=0 is seen by expanding in  $k_{eff}$ ,

$$e^{-\alpha t} = e^{-\frac{t}{\tau}} \left( 1 + \frac{k_{eff}}{\tau} t + \frac{1}{2} \left( \frac{k_{eff}}{\tau} \right)^{2} + \cdots \right)$$

$$= e^{-\frac{t}{\tau}} + \int_{0}^{t} e^{-\frac{(-t_{f})}{\tau} v} \frac{dt_{f}}{\tau} e^{-\frac{t_{f}}{\tau}} + \int_{0}^{t} e^{-\frac{(-t_{f2})}{\tau} v} \frac{dt_{f2}}{\tau} \int_{0}^{t_{f2}} e^{-\frac{(t_{f2}-t_{f1})}{\tau} v} \frac{dt_{f1}}{\tau} e^{-\frac{t_{f1}}{\tau}} + \cdots$$
A47

This is the probability that an initial neutron at t=0 survives until time t, plus the probability that a neutron created at t=0 survives until time  $t_f$ , and then between  $t_f$  and  $t_f+dt_f$  produces a

fission with probability  $\frac{dt_f}{t_f}$ , the fission produces  $\bar{v}$  neutrons, and these neutrons survive from

 $t_f$  until time t, and so on for a series of fissions following in time. Using  $\frac{\overline{v}}{\tau_f} = \overline{v} \frac{p}{\tau}$ , where

 $\frac{\tau}{\tau_f}$  = p, or  $\tau_f$  = p $\tau$ , the neutron lifetime against fission is longer than the total neutron lifetime

by the probability that the neutron induces fission, p. The average number of induced fissions is

$$\int_{0}^{\infty} e^{-\alpha t} \frac{dt}{\tau_{f}} = \frac{1}{\alpha \tau_{f}} = \frac{\tau}{\tau_{f}} \frac{1}{(1 - \nu p)} = \frac{p}{(1 - \nu p)} = \frac{M - 1}{\nu}$$
A48

where  $M = \frac{1}{(1-vp)}$  is the system multiplication (to be discussed below). The average number of

neutrons created by induced fissions is  $\nu$  times the average number of fissions, or M - 1. If M = 1, starting from one neutron at t = 0, there are no neutrons created by induced fission. We have shown in the text from solutions to Equation A53 below (and from Monte Carlo simulations) that the average number of created neutrons is a very poor characterization of the fission chain due to very large fluctuations in the number of neutrons created.

#### Fission chain neutron population

This analysis above for the internal neutron population can be generalized [9] to answer the question: how many neutrons were produced by the chain? We must keep track of those neutrons created that did not produce a subsequent fission. Some of the neutrons produced remain in the system at time t with a chance of perpetuating the chain, and some have left the system. Now we consider  $P_{k,m}(t)$ , the probability that starting from one neutron at t=0, there are k neutrons in the system at time t, that could perpetuate the chain, and m neutrons that were produced but leave the multiplying system by time t. The neutrons that leave the system may have been absorbed by non-fission processes, or leaked. Now

$$P_{k,m}(t + \Delta t) = P_{k,m}(t)\left(1 - k\frac{\Delta t}{\tau}\right) + qP_{k+1,m-1}(t)(k+1)\frac{\Delta t}{\tau} + p\sum_{\nu=0}P_{k-\nu+1,m}(t)C_{\nu}(k-\nu+1)\frac{\Delta t}{\tau}$$
 A49

This equation differs from Equation A32 by increasing m whenever a neutron disappears from the system, decreasing the number of neutrons in the system, and increasing the number of ends of branches of the fission tree. The probability generating function becomes a function of an extra variable,

$$f(t, x, y) = \sum_{k=0}^{\infty} \sum_{m=0}^{\infty} P_{k,m}(t) \chi^{k} y^{m}$$
 A50

The x variable keeps track of those neutrons in the system that can perpetuate the chain, while y keeps track of those neutrons that leave the system. The coefficients give the probabilities that at time t there are k neutrons in the system, having started from one neutron at t=0, *and* there are m neutrons that have been produced but are no longer in the multiplying system. The generating function now obeys the equation,

$$\tau \frac{\partial f}{\partial t} = g_{y}(x) \frac{\partial f}{\partial x}$$
 A51

which now depends on the extra parameter, y, through

$$g_{y}(x) = -x + qy + pC(x)$$
 A52

If y = 1, this equation reduces to the previous Equation A35. As seen from the generating function, Equation A50, the restriction to y = 1 sums the probabilities for the number of neutrons that were produced but have left the multiplying system. The analysis of the probability generating function equation is essentially the same as above for the internal neutron population since y is just a parameter as far as the differential equation is concerned. (The neutrons in the system create the evolving chain, the neutrons lost from the multiplying medium can no longer affect this evolution.) The  $t \to \infty$  limit is now characterized by the equation [7],

$$h(y) = qy + p \sum_{v \in \mathcal{V}} C_v \left[ h(y) \right]$$
 A53

where as  $t \to \infty$ ,

$$f(t, x, y) \rightarrow h(y)$$
 A54

The generating function is independent of x for the subcritical regime of interest.

#### **Exact Series Solution for General Asymptotic Chain Probability**

Equation A53 has a simple and elegant solution [9] using Lagrange's series formula [14] (further details on the derivation are given below):

$$h(y) = qy + \sum_{n_f=1}^{\infty} \frac{p^{n_f}}{n_f!} \left\{ \frac{d^n f^{-1}}{dz^n f^{-1}} C(z)^n f \right\}_{z=qy}$$

$$= qy + \sum_{n_f=1}^{\infty} \frac{p^{n_f}}{n_f!} \left\{ \frac{d^n f^{-1}}{dz^n f^{-1}} \left[ C_0 + C_1 z + C_2 z^2 + C_3 z^3 + \cdots \right]^n f \right\}_{z=qy}$$
A55

where  $C(z)^{n_f}$  has a natural multinomial expansion,

$$\left[ C_0 + C_1 z + C_2 z^2 + C_3 z^3 + \cdots \right]^{f} = \sum_{n_0, n_2, n_3, \cdots} \frac{n_f!}{n_0! n_1! n_2! \cdots} \left( C_0^{n_0} C_1^{n_1} C_2^{n_2} \cdots \right) z^{n_1 + 2n_2 + 3n_3 + \cdots}$$
A56

where the sums over  $n_0, n_1, n_2, \cdots$  are constrained,  $n_f = n_0 + n_1 + n_2 + \cdots$ . After the differentiations with respect to z in equation A55, the fission chain probability distribution [9] is determined from the coefficients of  $y^v$ ,

$$P_{v} = q \delta_{v,1} + q^{v} \sum_{n_{c}=1}^{\infty} \frac{p^{n_{f}} (n_{f} + v - 1)}{n_{f}!} \sum_{n_{0} \in n_{0} \in n_{0}, \dots} \frac{n_{f}!}{n_{0}! n_{1}! n_{2}! \dots} \left( C_{0}^{n_{0}} C_{1}^{n_{1}} C_{2}^{n_{2}} \dots \right)$$
 A57

where the sums over  $n_0, n_1, n_2, \cdots$  are doubly constrained: first  $n_f = n_0 + n_1 + n_2 + \cdots$ , which says that out of the  $n_f$  fissions,  $n_0$  of them produced no neutrons,  $n_1$  of the fissions produced 1 neutron,  $n_2$  of the fissions produced 2 neutrons, etc., and that the sum of fissions must add up to the total of  $n_f$  fissions. The second constraint is,  $n_f + v - 1 = n_1 + 2n_2 + 3n_3 + \cdots$ , which says that from the  $n_1$  fissions producing 1 neutron plus the  $n_2$  fissions producing 2 neutrons, etc., a total number of neutrons,  $n_f + v - 1$  were produced. Beginning with one neutron,  $n_f + v$  additional neutrons are produced,  $n_f$  of those produced were absorbed inducing fissions, and  $n_f + n_f + n_f$ 

#### Combinatorics of fission chain trees

The fission chain probability formula equation A56 is a sum over all possible trees. There is a remarkable simplification to the sum over all permutations of trees of specific topological classes [9], which we illustrate below. For each term with a specific number of final neutrons v there is a sum over the number of fissions,  $n_f$ . For a specific total number of fissions,  $n_f$ , there are further sums over the number of fissions that created a specific number of neutrons, where  $n_f = n_0 + n_1 + n_2 + \cdots$ . There are many trees with this constraint but the sum over the combinatorics of all possible trees with this constraint sums to a simple combinatorial factor,

$$\frac{(v + n_f - 1)}{v! n_0! n_1! n_2! \cdots}$$
 A58

As an example of this formula, the Figure A1 below shows a set of fission trees with v=6 neutrons created. They all have  $n_f=3$  fissions, 1 fission produced 2 neutrons, so  $n_2=1$ , and 2 fissions produced 3 neutrons, so  $n_3=2$ . The combinatorial factor is  $\frac{(v+n_f-1)}{v!n_2!n_3!} = \frac{(6+3-1)}{6!!!2!} = 28$ . The Figure A1 shows an enumeration of the 28 trees. There are permutation factors with each graph. For example, the bottom left graph has a permutation factor of 9: of the neutrons created by the first fission any of the 3 could induce the next fission, and any of the 3 from that fission could induce the final fission. The factor 28 is the sum of these permutations for all the graphs with the same values of v,  $n_f$ ,  $n_2$ ,  $n_3$ . The total contribution of these graphs is  $n_f^6$  and  $n_f^6$  and  $n_f^6$  are  $n_f^6$  are  $n_f^6$  and  $n_f^6$  are  $n_f^6$  and  $n_f^6$  are  $n_f^6$  are  $n_f^6$  and  $n_f^6$  are  $n_f^6$  are  $n_f^6$  and  $n_f^6$  are  $n_f^6$  and  $n_f^6$  are  $n_f^6$  and  $n_f^6$  are  $n_f^6$  and  $n_f^6$  are  $n_f^6$  are  $n_f^6$  and  $n_f^6$  are  $n_f^6$  and  $n_f^6$  are  $n_f^6$  and  $n_f^6$  are  $n_f^6$  are  $n_f^6$  and  $n_f^6$  are  $n_f^6$  and  $n_f^6$  are  $n_f^6$  are  $n_f^6$  and  $n_f^6$  are  $n_f^6$  are  $n_f^6$  and  $n_f^6$  are  $n_f^6$  are  $n_f^6$  and  $n_f^6$  are  $n_f^6$  are  $n_f^6$  and  $n_f^6$  are  $n_f^6$  and  $n_f^6$  are  $n_f^6$  and  $n_f^6$  are  $n_f$ 

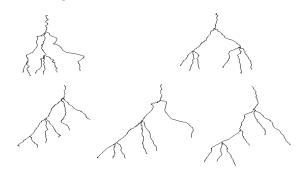


Figure A1 Starting from a single neutron, the topologically distinct fission chains are shown for  $n_f = 3$  fissions, of which:  $n_2 = 1$ ,  $n_3 = 2$ , and

 $n_f + v - 1 = 3 + 6 - 1 = 2 n_2 + 3 n_3 = 2 + 6$ . The six final neutrons arise from a total number of eight neutrons produced, of which three are absorbed in creating fissions, and there was one initial neutron. (If there were no fissions, there would still be one neutron.) Each of the graphs have associated combinatorial factors for the number of distinguishable permutations of the neutron lines. The first has 6 permutations, 3 distinguishable orderings of the single neutron branch, times two for the interchange of the two and three neutron branches. Similarly, the second tree has 1, the third has 9, the fourth 6, and the last 6 permutations. The total number of permutations is then, 6+1+9+6+6=28.

Another term of the series, also with  $n_f = 3$  fissions and v = 6 ends of the chains, this time with one of the fissions producing 4 neutrons, and two of the fissions producing 2 neutrons, is shown in the Figure A2 below. This set of four topologically distinct graphs occurs with total probability  $q^6 p^3 28 C_2^2 C_4$ .

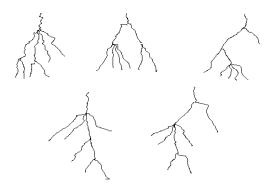


Figure A2 Starting from a single neutron, the topologically distinct fission chains are shown for  $n_f = 3$  fissions, of which:  $n_2 = 2$ ,  $n_4 = 1$ , and  $n_f + v - 1 = 3 + 6 - 1 = 2 n_2 + 4 n_4 = 4 + 4$ . Each of the graphs have associated combinatorial factors for the number of distinguishable permutations of the neutron lines. The first has 6 permutations, the second tree has 2, the third has 4, the fourth 8, and the last 8 permutations. The total number of permutations is then, 6+2+4+8+8=28.

There are additionally many other terms with  $q^6 p^3$  and  $n_f = n_0 + n_1 + n_2 + \dots + n_6 = 3$  and  $n_f + v - 1 = n_1 + 2n_2 + 3n_3 + \dots + 6n_6 = 6$ .

#### Multiplication and fission trees

The Lagrange series representation for the fission chain also gives a precise relation between system multiplication and a discrete counting method [9]. The examples of Figures A1 and A2 above show the precise relation between, on one hand, counting nodes and branches, and on the other hand, counting leaves. The ends of the lines are leaves, representing the neutrons we wish to track, of number v, the nodes are the fissions, of number  $n_f$ , and the branches, b, are the number of neutrons created by fission processes (with probabilities  $n_f$ . In the examples of Figures A1 and A2 each graph has  $n_f$  = 3 nodes, or fissions, and the number of neutrons produced by the fissions,  $n_f$  = 8. The number of branches minus nodes plus one, for the initial neutron, equals the number of leaves,  $n_f$  = 8 - 3 + 1. In general,

$$v = b - n_f + 1$$
 A59

Using a Monte Carlo simulation, for example, one could simulate fission chains and determine the probability distribution,  $P_{\nu}$ , to create  $\nu$  neutrons. For each initial neutron, one could tally the number of fissions, and the number of neutrons produced by those fissions; Equation A59 would then give the number of neutrons at the ends of the chain. The relation between this probability distribution,  $P_{\nu}$ , and the parameter, p, the probability that a neutron produced by

fission induces a subsequent fission, can be determined from the first moment. From the definition, Equations A50 and A54,

$$h(y) = \sum_{v=0}^{\infty} P_v y^v$$
 A60

Differentiating Equation A60 with respect to y gives

$$h'(y) = \sum_{v=1}^{\infty} v P_v y^{v-1} = q + pC'[h(y)]h'(y)$$
 A61

Setting y = 1, h(1) = 1 since  $P_v$  is a normalized probability distribution,  $C'[1] = \overline{v}$ , the average number of neutrons emitted in an induced fission, as seen from Equation A44. Then

$$\sum_{v=1}^{\infty} v P_v = \frac{q}{1 - vp}$$
 A62

The expression on the right is usually referred to as *escape multiplication*, the expression without the q factor is the total multiplication, and counts the extra  $n_f$  neutrons absorbed to induce fissions.

#### Moments of the counting distribution

The fission chain probability distribution,  $P_v$  enters linearly into the  $\Lambda_j$  (from Equation A19 or A27), the probability that j neutrons out of the chain are detected within the time gate T, but the  $\Lambda_j$  enter the count distribution  $b_n(T)$  as powers (Equation A15). (In the generating function for the count distribution the  $\Lambda_j$  enter linearly in the exponent, Equation A18.) The combinatorial moments of the counting distribution are, however, linear in moments of  $P_v$ , as well as linear in the fission rate. Consider first the combinatorial moments of the  $\Lambda_j$  [5],

$$Y_{q}(T) = \sum_{j=q}^{\infty} {j \choose q} \Lambda_{j}(T)$$
 A63

The  $Y_q$  will be seen to be cumulant moments of the counting distribution, and give the probability of detecting q neutrons with a common ancestor. This common ancestor is a fission event at some point in the fission chain. All higher  $\Lambda_j$  with  $j \ge q$  contribute to this probability. While  $\Lambda_j$  is the probability to detect j neutrons within the time gate T form the  $\nu$  emitted in the

chain, there are  $\binom{j}{q}$  ways that these j neutrons could contribute to q. For example,

$$Y_3(T) = \Lambda_3(T) + 4\Lambda_4(T) + 10\Lambda_5(T) + \cdots,$$
 A64

the probability to detect, within the time gate T, 3 neutrons with a common ancestor and is the probability  $\Lambda_3(T)$  that 3 neutrons out of the  $\nu$  emitted in the chain are detected within time gate T, plus 4 ways that 3 neutrons could have a common ancestor if 4 neutrons out of the  $\nu$  emitted

in the chain are detected within time gate T, plus 10 ways that 3 neutrons could have a common ancestor if 5 neutrons out of the  $\nu$  emitted in the chain are detected within time gate T, etc. In order to extract these probabilities from the measured data, first combinatorial moments of the counting distribution are required,

$$M_{q} = \sum_{n=q}^{\infty} \binom{n}{q} b_{n}(T)$$
 A65

These moments are the probabilities of detecting q multiplets if n neutrons were detected within the time gate T. For example,  $M_2$  is the sum of probabilities for all the ways that a pair of neutrons could be detected, if 2 or more neutrons were counted within the time gate T. These moments can be computed from the generating function Equation A18 for the generalized Poisson distribution [5]; the derivatives of the generating function give the factorial moments of the counting distribution when expanded about z = 1 (expanding about z = 0 gives the number of counts). The first two moments, from the first two derivatives of Equation A18, are:

$$\left[\frac{d\pi}{dz}\right]_{z=1} = \sum_{n=1}^{\infty} n b_n = \left[\left(\sum_{j=1}^{\infty} j \Lambda_j z^{j-1}\right) e^{\sum_{j=1}^{\infty} \left(z^{j-1} \Lambda_j\right)}_{z=1} = \sum_{j=1}^{\infty} j \Lambda_j \quad A66$$

$$\frac{1}{2} \left[ \frac{d^2 \pi}{dz^2} \right]_{z=1} = \sum_{n=2}^{\infty} \frac{n(n-1)}{2} b_n = \sum_{j=2}^{\infty} \frac{j(j-1)}{2} \Lambda_j + \frac{1}{2} \left( \sum_{j=1}^{\infty} j \Lambda_j \right)^2$$
 A67

Out of all the pairs, there is an uncorrelated piece where one neutron could come from one fission chain and the other neutron could come from an independent chain, as well as the correlated probability that both neutrons could arise from the same chain, (from Equations A67, A65 and A63),

$$M_2 = \frac{\overline{C}^2}{2} + Y_2$$
 A68

where  $\overline{C} = M_1$  is the probable number of counts within the time gate T,

$$\overline{C} = \sum_{n=1}^{\infty} n b_n = b_1 + 2 b_2 + 3 b_3 + \dots = R_1 T$$
 A69

 $R_1$  is the count rate. Similarly, the fully correlated part of the third moment is,

$$Y_{3} = \sum_{n=3}^{\infty} {n \choose 3} b_{n}(T) - Y_{2}\overline{C} - \frac{\overline{C}^{3}}{3!}$$
 A70

Out of all possible triples of counts, the probability that a pair of those counts has a common ancestor and the third is from an independent chain, and the probability that all three are from independent chains, must be subtracted in order to get the probability that all three neutrons were from the same chain.

The explicit form for  $Y_q(T)$  is obtained by applying the identity,

$$\sum_{b=c}^{a} {a \choose b} {b \choose c} x^b (1-x)^{a-b} = {a \choose c} x^c$$
 A71

to the expression for  $\Lambda_j(T)$ , Equation A21, in Equation A63 for  $Y_q(T)$ . This leads to the striking simplification,

$$Y_{q}(T) = F_{s} \sum_{v=q}^{\infty} P_{v} {v \choose q} \varepsilon^{q} \left[ \int_{-\infty}^{0} x^{q} ds + \int_{0}^{T} y^{q} ds \right]$$
 A72

where x and y are given in Equations A22 and A23. The integrals over the fission time s become,

$$\int_{-\infty}^{0} \chi^{q} ds = \left(1 - e^{-\lambda T}\right) \int_{-\infty}^{0} e^{q\lambda s} ds = \left(1 - e^{-\lambda T}\right) \frac{1}{\lambda q}$$
 A73

and

$$\int_{0}^{T} y^{q} ds = \int_{0}^{T} \left( 1 - e^{-\lambda (T - s)} \right) ds$$
 A74

For q = 1, Equations A72 - A74 give,

$$\overline{C} = \varepsilon F_s \left[ \sum_{v=1}^{\infty} \mathbf{v} P_v \right] T = \varepsilon q M \overline{\mathbf{v}}_s F_s T$$
 A75

The average number of neutrons created by a fission chain initiated by a single neutron is given by Equation A62; when there is spontaneous fission there are  $\overline{\boldsymbol{v}_s}$  initiating neutrons on average, where  $\overline{\boldsymbol{v}_s}$  has the same relation to the spontaneous fission distribution as Equation A44 for induced fission. For q = 2, this gives,

$$Y_{2} = \varepsilon^{2} F_{s} \left[ \sum_{\nu=2}^{\infty} P_{\nu} {v \choose 2} \right] T \left( 1 - \frac{1 - e^{-\lambda T}}{\lambda T} \right)$$
 A76

This equation can also be derived by a different method [1 and 3], starting from pair correlation functions, the probability that if a neutron is detected between  $t_1$  and  $t_1 + dt_1$ , a second neutron is detected between  $t_2$  and  $t_2 + dt_2$ . Integrating the correlation function over all pairs of times within the counting time gate gives Equation A75. Equation A76 is linear in the spontaneous fission rate, linear in (a moment of) the fission chain number distribution, and has factored the efficiency dependence. This factored dependence is in contrast to the way these parameters enter nonlinearly or convolved in the count distribution, Equations A15 and A19.

The moments of the fission chain can be obtained from the general series expansion of a probability generating function,

$$f(x) = \sum_{n=0}^{\infty} P_n x^n = \sum_{n=0}^{\infty} \left[ \sum_{\nu=n}^{\infty} P_{\nu} {v \choose n} \right] (x-1)^n$$
 A77

The factorial moments of the fission chain distribution are obtained by expanding the fission chain generating function about x = 1. By differentiating the fission chain generating function  $C^{spont}[h(y)]$ , which generates the fission chain initiated by spontaneous fission, where  $C^{spont}(x)$  is the generating function for the spontaneous fission neutron distribution (like Equation A36 for the induced fission distribution), and h(y) satisfies Equation A53, and using the functional chain rule, we obtain [5]:

$$\sum_{v=2}^{\infty} P_v {v \choose 2} = \left(\frac{q}{1-vp}\right)^2 \left[v_{2s} + \frac{p}{1-vp}v_{s}v_{2}\right]$$
 A78

where  $\bar{v}$  defined above Equation A42, and  $v_2 = \sum_{v=2}^{\nu} {v \choose 2} C_v$  are moments of the induced fission

neutron number distribution, and  $\overline{\boldsymbol{V}}_s$  and  $\boldsymbol{V}_{2s}$  are the corresponding moments of the spontaneous fission distribution. From Equation A48, the factor,

$$\frac{p}{1-\overline{v}p}\overline{v}_s = \frac{M-1}{\overline{v}}\overline{v}_s$$
 A79

is the average number of fissions in a chain initiated by spontaneous fission. Equation A78 is the total number of ways that a pair of neutrons can have a common ancestry within the fission chain, they can come form one neutron from each of a pair of chains arising from the spontaneous fission, or the pair can arise from any of the induced fissions in the total chain. Equation A78 inserted into Equation A76 gives a formula for the Feynman variance [1].

Similarly for q=3, we obtain [5]:

$$Y_3 = \varepsilon^3 F_s \left[ \sum_{\nu=3}^{\infty} P_{\nu} {v \choose 3} \right] T \left( 1 - \frac{3 - 4e^{-\lambda T} + e^{-2\lambda T}}{2\lambda T} \right)$$
 A80

This equation can also be derived using the correlation function arguments illustrated in [3]. The number of ways to make a triple of neutrons out of v created by the chain is [5],

$$\sum_{v=2}^{\infty} P_v {v \choose 3} = \left(\frac{q}{1-\overline{v}p}\right)^3 \left(v_{3s} + \frac{p}{1-\overline{v}p} \left(\overline{v}_{s} v_3 + 2v_{2s} v_2\right)\right) + \left(\frac{p}{1-\overline{v}p}\right)^2 2\overline{v}_{s} v_2^2\right)$$
 A81

There are 4 topologically distinct ways that three of the neutrons can have common ancestry within the fission chain. One is that one neutron could come from each of three separate chains arising from the spontaneous fission. Another triple can arise from a single induced fission in any of the separate spontaneous fission chains. There could be a pair of spontaneous fission chains with one of the induced fissions in one of them being a common ancestor of a pair (there are 2 permutations of this possibility, either chain coming from the spontaneous fission could create

the induced fission that is a common ancestor of a pair). Finally any of the spontaneous fission chains could create an induced fission pair of chains, one of these induced fission chains having another induced fission that is the common ancestor of a pair (again there are 2 permutations).

#### Derivation of Lagrange series for fission chain distribution

We give more details on the derivation of the Lagrange series expansion solution of Equation A53. (Lagrange invented this series to solve Kepler's equation for the time evolution of an elliptical orbit,  $E = M + e \sin(E)$ , where E is the eccetric anomaly, the angle of the planet from the center of the ellipse, M is the mean anomaly  $(2\pi/P)$  t, where P is the orbit period, and e is the eccentricity. With the sine function expressed as a series, this equation for E(M) is of the same form as the fission chain formula for h(y). The Lagrange formula is an inversion of the series for M as a series in E, to give E as a series in M.) We will use a method from complex variable theory. This equation for the time asymptotic fission chain generating function can be written as the zero of the equation,

$$f(h) = h - qy - pC(h)$$
 A82

The *generalized argument theorem* of complex variable theory [14] gives h in terms of a contour integral over h that contains within the contour one simple zero of f(h),

$$h = \frac{1}{2\pi i} \oint z \frac{f'(z)}{f(z)} dz$$
 A83

Applying this theorem to f(h)of Equation A82,

$$h(y) = \frac{1}{2\pi i} \oint z \frac{(1 - pC'(z))}{z - qy - pC(z)} dz$$

$$= \frac{1}{2\pi i} \oint z \frac{1 - pC'(z)}{z - qy} \frac{1}{1 - \frac{p}{z - qy}} C(z) dz = \frac{1}{2\pi i} \oint z \frac{1 - pC'(z)}{z - qy} \sum_{n_f=0}^{\infty} \frac{p^{n_f}}{(z - qy)^{n_f}} C(z)^{n_f} dz \quad A84$$

Expanding the product of (1 - p C'(z)) and the sum, separating the  $n_f = 0$  term in the first sum, and using Cauchy's theorem, gives,

$$= \frac{1}{2\pi i} \oint \left\{ \frac{z}{z - qy} \left[ 1 + \sum_{n_{f}=1}^{\infty} \frac{p^{n_{f}}}{(z - qy)^{n_{f}}} C(z)^{n_{f}} \right] - \frac{z}{z - qy} pC'(z) \sum_{n_{f}=0}^{\infty} \frac{p^{n_{f}}}{(z - qy)^{n_{f}}} C(z)^{n_{f}} \right\} dz$$

$$= qy + \frac{1}{2\pi i} \sum_{n_{f}=1}^{\infty} p^{n_{f}} \oint \left\{ \frac{z}{(z - qy)^{n_{f}+1}} C(z)^{n_{f}} - \frac{z}{(z - qy)^{n_{f}}} C'(z) C(z)^{n_{f}-1} \right\} dz \quad A85$$

In the last term the summation on  $n_f$  was shifted to begin at 1, absorbing the extra factor of p in front of C'(z). The terms in curly brackets of the last term of Equation A85 can be regrouped as a derivative, and using Cauchy's theorem for derivatives,

$$= qy + \frac{1}{2\pi i} \sum_{n_f=1}^{\infty} \frac{p^{n_f}}{n_f} \int z \frac{d}{dz} \frac{C(z)^{n_f}}{(z - qy)^{n_f}} dz = qy + \sum_{n_f=1}^{\infty} \frac{p^{n_f}}{n_f} \frac{d^n f^{-1}}{dz^n f^{-1}} C(z)^n f \bigg|_{z=qy}$$
A86

This gives Equation A55. With Equation A56 for the multinomial expansion,

$$h = qy +$$

$$\sum_{n_{f}=1}^{\infty} \frac{p^{n_{f}}}{n_{f}} \frac{d^{n_{f}-1}}{d_{z}^{n_{f}-1}} \left[ \sum_{n_{0},n_{1},n_{2},\cdots} \frac{n_{f}!}{n_{0}! n_{1}! n_{2}! \cdots} \left( C_{0}^{n_{0}} C_{1}^{n_{1}} C_{2}^{n_{2}} \cdots \right) \right]_{z=av}^{n_{1}+2n_{2}+3n_{3}+\cdots}$$
A87

Defining v from the condition,

$$n_f - 1 + v = n_1 + 2n_2 + 3n_3 + \cdots$$
 A88

the  $n_f$ -1 derivatives acting on z give,

$$\frac{d^{n_{f}^{-1}}}{dz^{n_{f}^{-1}}} z^{n_{1}+2n_{2}+3n_{3}+\cdots}$$

$$= \sum_{v=0}^{\infty} \delta_{n_{f}^{-1+v},n_{1}+2n_{2}+3n_{3}+\cdots}} \left(n_{f}^{-1+v}\right) \left(n_{f}^{-2+v}\right) \left(n_{f}^{-3+v}\right) \cdots \left(v+1\right) z^{\left(n_{f}^{-1+v}\right) \left(n_{f}^{-1}\right)}$$

$$= \sum_{v=0}^{\infty} \delta_{n_{f}^{-1+v},n_{1}+2n_{2}+3n_{3}+\cdots}} \frac{\left(n_{f}^{-1+v}\right) \left(n_{f}^{-1+v}\right)}{v!} z^{v}$$
A89

Finally, Equations A87 and A89 give the main result, Equation A57, in the generating function form,

$$h(y) = qy + \sum_{v=0}^{\infty} \left[ q^{v} \sum_{n_{f}=1}^{\infty} \frac{p^{n_{f}}}{n_{f}!} \frac{(n_{f}+v-1)}{v!} \sum_{n_{0},n_{1},n_{2},\cdots} \frac{n_{f}!}{n_{0}! n_{1}! n_{2}! \cdots} \left( C_{0}^{n_{0}} C_{1}^{n_{1}} C_{2}^{n_{2}} \cdots \right) \right] y^{v} A90$$

subject to the constraints on the sums over  $n_0, n_1, n_2, \cdots$ ,  $n_f + v - 1 = n_1 + 2n_2 + 3n_3 + \cdots$ , from Equation A89, and  $n_f = n_0 + n_1 + n_2 + \cdots$ , from the multinomial expansion, Equation A56.

# Summary of neutron counting distribution for passive counting, and active interrogation with an $(\alpha, n)$ source

The final results for the count distribution produced by a fission source are summarized here. We will include in the formulas fission chains initiated by spontaneous fission and by  $(\alpha, n)$  sources, as well as an external random source. The external source shines directly on the detectors as well as inducing fission chains. The generating function for the count distribution has the form

$$\sum_{n=0}^{\infty} b_n(T) y^n = \exp \left[ R^{ext} T(y-1) \right]$$

$$+ \left[ S_{\alpha} + S^{ind} \left( S^{ext} \right) \right] \left[ \frac{1}{(-e^{-\lambda t})} \left[ \left( 1 - y \right) \left( 1 - e^{-\lambda t} \right) \right] - 1 \right] t$$

$$+ F_s \int_0^T \frac{1}{(-e^{-\lambda t})} \left[ C^{spont} \left[ h \left[ 1 - \varepsilon \left( 1 - y \right) \left( 1 - e^{-\lambda t} \right) \right] - 1 \right] dt \right]$$
A91

The first term in the exponent of generating function describes the direct shine contribution from external random source. It's contribution depends only on the count rate,  $R^{ext}$ , which is a product of the actual source strength,  $S^{ext}$ , and the probability to detect one of the neutrons created by the external source, an efficiency parameter. The second term describes fission chains initiated by a single neutron, either an  $(\alpha, n)$  source or a source induced by the external random source. The magnitude of the external source depends on the external source strength and on geometry that determines the induced fission probability. The last term is due to fission chains initiated by spontaneous fission. The generating function  $C^{spont}(x)$  is the  $\chi^v$  weighted sum of

spontaneous fission probabilities,  $C^{spont}_{v}$  to create specific numbers of neutrons, analogous to Equation A36 for the induced fission probabilities. The three terms in the exponent of Equation A91 will be discussed separately below.

The external random source contributes to every specific number of counts and every time gate in a way that depends on only a single parameter,  $R^{ext}$ . Because the form of its contribution is precisely known, the optimization of the comparison of experimental data and theory can determine this contribution robustly. It is this principle that enables any precisely understood distribution (such as eventually the cosmic ray distribution) to be separated from the contribution of the HEU.

The count distribution again has the form,

$$b_n(T) = \left(\Lambda_n + \Lambda_{n-14}\Lambda_1 + \Lambda_{n-2}\Lambda_2 + \Lambda_{n-2}\frac{\Lambda_1^2}{2} + \dots + \frac{\Lambda_1^n}{n!}\right)b_0$$
 A92

where  $b_0$  is given in Equation A16. The  $\Lambda_n(T)$  are linear in the different sources. The count distribution is of course nonlinear in the  $\Lambda$ 's. The  $\Lambda_n(T)$  are determined form the coefficient of  $y^n$  arising from expanding the *exponent* of Equation A91 in powers of y about y = 0. For  $(\alpha, n)$  sources,  $\Lambda_n(T)$  is,

$$\Lambda_{n}(T) = S_{\alpha} \int_{0}^{T} \left( 1 - e^{-\lambda t} \right)^{-1} \sum_{\nu=n}^{\infty} P_{\nu} {v \choose n} \varepsilon^{n} \left[ 1 - \varepsilon \left( 1 - e^{-\lambda t} \right) \right]^{-n} dt$$
A93

In the limit  $T >> \lambda^{-1}$  the factors of  $e^{-\lambda t}$  can be neglected, and the integral simplifies to a form easy to interpret,

$$\Lambda_{n}(T) \to S_{\alpha} T \sum_{\nu=n}^{\infty} P_{\nu} \binom{\nu}{n} \varepsilon^{n} (1 - \varepsilon)^{\nu-n}$$
 A94

The factor  $S_{\alpha}T$  is the probable number of  $(\alpha, n)$  reactions that can initiate a fission chain within the time gate T. The  $P_{\nu}$  is the probability that  $\nu$  neutrons were created by the chain, and  $\binom{\nu}{n} \varepsilon^n (1-\varepsilon)^{-n}$  is the probability to count n neutrons out of the  $\nu$  created, if detecting a single

neutron has probability  $\varepsilon$ . For shorter time gates T, the factors of  $(-e^{-\lambda t})$  are giving a time dependent efficiency, lower for short times while the neutrons are independently diffusing before being counted. This is explicitly seen in Equation A24, that separates the two contributions of chains, from those initiated during the time gate and chains initiated before the opening of the time gate.

The spontaneous fission term is a convolution,

$$C^{spont} \left[ h \left[ 1 - \varepsilon (1 - y) \left( 1 - e^{-\lambda t} \right) \right] \right] = \sum_{v=0}^{\infty} C^{spont} \left( h \left[ 1 - \varepsilon \left( 1 - y \right) \left( 1 - e^{-\lambda t} \right) \right] \right)$$
 A95

The spontaneous fission can create some number of neutrons from 0, 1, 2,..., typically up to about 7. Each of these neutrons can create a chain, and thus the power of the h distribution that generates a chain from a single neutron (Equation A60 with the explicit form Equation A90). The fission chain generating function variable in h(z) is replaced by the time dependent function,  $z \to 1 - \varepsilon (1 - y) (1 - e^{-\lambda z})$ , (as derived from Equations A19 to A31).

#### Fission chain γ rays

The fission chain rate equation can also keep track of the  $\gamma$  rays emitted by individual fission in the chain. The rate equation is

$$P_{k,m,s}(t + \Delta t) = P_{k,m,s}(t) \left( 1 - k \frac{\Delta t}{\tau} \right)$$

$$+ q P_{k+1,m-1,s}(t) (k+1) \frac{\Delta t}{\tau} + p \sum_{v=0} P_{k-v+1,m,s-v}(t) C_v G_v (k-v+1) \frac{\Delta t}{\tau}$$
A96

In this equation, in the second term on the right hand side we make the simplifying assumption that when a neutron is lost from the chain it is by leakage and not absorption or other reactions that create  $\gamma$  rays. (Because the neutron capture cross sections are small compared to fission for fission spectrum neutron energies, we neglect them here. Generally, though, the neutron capture processes  $(n, \gamma)$  create cascades of  $\gamma$  rays, like fission, for example sub-keV neutron capture in  $^{238}$ U. The number of  $\gamma$  rays are changed by fission in the last term. The fission is induced by one of the (k - v + 1) neutrons in the system before the fission, absorbing 1 neutron and creating v neutrons with probability  $C_v$ . The same fission creates  $\gamma$  additional  $\gamma$  rays with probability  $G_{\gamma}$ .

The probability generating function for the time dependent populations is now,

$$f(t, x, y, w) = \sum_{k=0}^{\infty} \sum_{m=0}^{\infty} \sum_{s=0}^{\infty} P_{k,m,s}(t) \chi^{k} y^{m} w^{s}$$
 A97

The generating function satisfies the equation

$$\tau \frac{\partial f}{\partial t} = \left(-x + qy + pG(w)C(x)\right)\frac{\partial f}{\partial x}$$
 A98

This equation is similar to the rate equation for the internal neutron population, Equations A51 and A52, and is solved in the same way. The asymptotic fission chain distribution as  $t \to \infty$  for the number of created neutrons and  $\gamma$  rays,

$$f(t, x, y, w) \rightarrow Q(y, w)$$
 A99

satisfies the functional equation,

$$Q(y,w) = qy + pG(w) \sum_{v=0} C_v [Q(y,w)]^v$$
 A100

The solution for the generating function is the same as Equation A90 but with  $p^{n_f \epsilon}$  replaced by  $[pG(w)]^{n_f}$ ,

$$Q(y,w) = qy + \sum_{v=0}^{\infty} \left[ (qy) \sum_{n_{f}=1}^{\infty} \frac{\left[ G(w)p \right]^{n_{f}}}{n_{f}!} \frac{(n_{f}+v-1)}{v!} \sum_{n_{0},n_{1},n_{2},\cdots} \frac{n_{f}!}{n_{0}!n_{1}!n_{2}!\cdots} \left( C_{0}^{n_{0}} C_{1}^{n_{1}} C_{2}^{n_{2}}\cdots \right) \right]$$
A101

with the same constraints on the sums over  $n_i$  given below Equation A90.

#### Gamma ray counting distribution

The generating function for the  $\gamma$  ray counting distribution is

$$\pi(w,y,z) = \sum_{k=0}^{\infty} \sum_{m=0}^{\infty} \sum_{n=0}^{\infty} b_{k,m,n}(T) w^{k} y^{m} z^{n}$$

$$= \exp \left[ F_{s} \int \left\{ G_{s} \left[ -\varepsilon_{\gamma} (1-w) \right] C^{spont} \left[ \varrho \left[ -\varepsilon (1-y) (1-e^{-\lambda t}) \right] - \varepsilon_{\gamma} (1-z) (1-e^{-\lambda t}) \right] - \varepsilon_{\gamma} (1-y) \left[ -e^{-\lambda t} \right] \right\} dt$$

$$+ F_{s} \int_{0}^{T} \frac{e^{-\lambda t}}{(-e^{-\lambda t})} \left\{ C^{spont} \left[ h \left[ -\varepsilon (1-y) (1-e^{-\lambda t}) - \varepsilon_{\gamma} (1-z) (1-e^{-\lambda t}) \right] \right] - 1 \right\} dt$$

$$+ F_{s} \sum_{k=0}^{T} \int_{0}^{T} \frac{e^{-\lambda_{\beta_{k}} t}}{(-e^{-\lambda_{\beta_{k}} t})} \left\{ C^{spont} \left[ F \left[ G^{\beta_{k}} \left[ 1-\varepsilon_{\gamma} (1-w) \right] \right] - 1 \right] dt$$

$$+ R^{\alpha} T(w-1) - A 102$$

where the generating function Q(y, w) for the number of neutrons (tracked by y) and  $\gamma$  rays (tracked by w) created by a chain satisfies Equation A100, and where the generating function for the number of fissions in a chain F(x) is

$$F(x) = q + pC[F(x)]$$
 A103

The generating function  $G_s(x)$  is for the probability distribution for the number of  $\gamma$  rays created by spontaneous fission. The generating function  $C^{spont}(x)$  is the probability distribution for the number of spontaneous fission neutrons. The generating function  $\mathbf{h}(x)$  is the probability distribution for the number of neutrons created by a chain initiated by a single neutron, and satisfies Equation A90. The generating function  $G^{\beta_i}(x)$  is the probability distribution for the number of cascade  $\gamma$  rays created by the  $\beta$  decay of the  $i^{th}$  wave of fission fragments. The rate  $R^{\alpha}$  is of  $\gamma$  rays emitted in the  $\alpha$  decay chains.

The first term in the exponent of the count distribution generating function, Equation A102, is from those fission chains initiated during the time gate T. There are  $\gamma$  rays emitted in the

spontaneous fission, and the fission chain emits different numbers of neutrons and  $\gamma$  rays. The neutrons are detected after a diffusion time scale  $\chi^{-1}$ . There are also  $\gamma$  rays created by neutron capture on the same time scale. The time dependence of this term is derived as the last term of Equation A24.

The second term of Equation A102 is from fission chains initiated before the time gate is opened. The neutrons are diffusing on a time scale  $\lambda^{-1}$  and can be detected during the time gate even though the chain created them before the time gate opened. Similarly there are  $\gamma$  rays created by neutron capture after diffusion.

The third term of Equation A102 is from the  $\beta$  decay of fission fragments. The fission fragments were created by chains seconds before the opening of the time gate. There are multiple  $\beta$  decays of the fission fragments producing different distributions of cascade  $\gamma$  rays. The time dependence of the second and third terms of Equation A102 was derived in the first term of the last line of Equation A24 and transformed to the current form in Equation A26.

The last term generates a Poisson distribution due to the (strong) randomly created  $\gamma$  rays from the  $\alpha$  decay chains. (We neglect for simplicity the few  $\gamma$  cascades in the  $\alpha$  decay chains. We also neglect the small contribution of  $\beta$  delayed neutron emission, which gives a source of the same form as  $(\alpha, n)$  reactions, sometimes even with accompanying  $\gamma$  rays. Even the  $\gamma$  ray cascades from fission fragment  $\beta$  decay have not yet been exploited.)

#### Time interval distribution

The time data stream of counts can be analyzed in different ways. One of the fundamental characterizations of the time series is the probability distribution for time intervals between adjacent counts. This distribution is particularly useful for multiplying HEU which has rare fission chains. The initiation of chains is random, but following a random initiation is a relatively closely spaced series of counts from the fission chain, that are spread out in time due to neutron diffusion. For neutron counting there are two important time scales, one associated with the initiation rate of chains, and the other with the neutron diffusion spread of promptly created neutrons from the fission chain. The actual time intervals also depend on detection efficiency and on multiplication.

For a random source, the probability there is no count between time t = 0 and time t, and then a count between t and  $t + \Delta t$  is described above Equation A13. This implies that the probability for a time interval t between counts is proportional to  $e^{-Rt}$ , where again R is the count rate. This says that probability is highest to get another count soon after the initial count, but that it is possible to have a long time between counts. As we saw in Equation A14, the average time between counts is  $R^{-1}$ . In HEU the time interval between the initiation of fission chains is very long, say 0.1 second. Following the random initiation event there is a fission chain, and there could then be several counts following in time on a 10  $\mu s$  time scale characteristic of neutron diffusion. The probability for the time interval T between counts now has two terms associated with adjacent counts being from the same fission chain, and adjacent counts being between the last count of a chain and the first count of the next chain. The formula is,

$$I_0(T)\Delta T = R_1 \Delta T r_0 n_0 + \lambda \Delta T \sum_{n=2}^{\infty} \frac{e_n(\varepsilon)}{N} \left( \sum_{k=0}^{n-1} k e^{-k\lambda T} \right) b_0(T)$$
 A104

In this equation,  $R_1$  is the count rate for a multiplying system,

$$R_1 = \varepsilon q M \overline{\mathbf{v}}_s F_s$$
 A105

The fission rate is  $F_s$ , on average there are  $\overline{V}_s$  neutrons created by the spontaneous fission, the system multiplies the initial neutrons with multiplication M which is the average number of neutrons created by fission chains initiated by a single neutron, q M is the escape multiplication,  $M - \frac{M-1}{\overline{V}}$ , the subtracted term being the average number of induced fissions (that each take a neutron from the chain), and  $\varepsilon$  is the probability to count a neutron created by the fission chain;

$$e_n(\varepsilon) = \sum_{\nu=0}^{\infty} P_{\nu} \binom{\nu}{n} \varepsilon^n (1 - \varepsilon)^{-n}$$
 A106

is the probability to count n neutrons from a fission chain that created v neutrons (excluding those internally absorbed to create subsequent fissions);

$$r_0 = \sum_{n=1}^{\infty} \frac{e_n(\varepsilon)}{N} \left( \sum_{k=0}^{n-1} e^{-k\lambda T} \right)$$
 A107

is the probability that no neutrons are detected from the same fission chain a time T after the initial trigger count [6], and where N is a normalization factor so that r<sub>0</sub> goes to 1 as T goes to zero,

$$N = \sum_{n=1}^{\infty} n \, \boldsymbol{e}_n(\varepsilon) = \varepsilon q M \overline{\boldsymbol{v}_s}$$
 A108

The probability to get no counts in a random time interval T is [9],

$$b_0(T) = \exp\left[-F_s \int_0^T \frac{1}{\left(-e^{-\lambda t}\right)} \left\{1 - \sum_{v=0}^{\infty} P_v \left[1 - \varepsilon \left(1 - e^{-\lambda t}\right)\right]\right\} dt\right]$$
 A109

and the probability for no count for a time T after a trigger count is [6],

$$n_0(T) = r_0(T) b_0(T)$$
 A110

This is the probability that there is no count for a time T after the trigger.

The first term of Equation A104 for  $I_0(T)$   $\Delta T$  contains the probability for the time interval T between the last neutron count of a chain and the first count of the next chain. For HEU that has rare chains,  $T >> \lambda^{-1}$ , so this term is approximately, from Equation A107, where only the k = 0 term survives, and Equation A106, which is a normalized probability distribution, and Equation A109,

$$R_1 \Delta T r_0 n_0 \approx R_1 \Delta T \frac{1}{N^2} \exp \left[ -F_s T \left\{ 1 - \sum_{v=0}^{\infty} P_v \left( 1 - \varepsilon \right)^v \right\} \right]$$
 A111

This is proportional to a Poisson time interval distribution with count rate  $F_s \left\{ 1 - \sum_{v=0}^{\infty} P_v \left( 1 - \varepsilon \right)^v \right\}$ . The first term of Equation A104 also describes time intervals where a neutron from a different chain is counted before another neutron from the same chain.

where a neutron from a different chain is counted before another neutron from the same chain. This probability is small for HEU with rare initiation events.

The second term of  $I_0(T)$   $\Delta T$ , Equation A104, is the probability for a time interval T between counts from neutrons from the same fission chain. For HEU, these time intervals T <<  $F_s^{-1}$ , and are separated by a time scale proportional to  $\lambda^{-1}$ .

The formula for  $I_0(T)$ , Equation A104, is derived [15] by both a combinatorial enumeration of processes and by a more formal relation between probability density distributions and cumulative distributions. The formal method could be immediately generalized to the next and subsequent neighbor distributions.